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Abstract

A set of approximate first-order ordinary differential equations was derived from the general dynamic equation (GDE) to study the aerosol dynamics with homogeneous nucleation and condensational diffusion-limited growth in an isothermal batch reactor. A lognormal size distribution was used, as an example, to illustrate the closure of the problem associated with the recursive nature of the moment equations in the formulation. The resulting set of approximate equations can be solved numerically to obtain the temporal variations of number concentration, volume fraction, vapor depletion, and other moments.

Keywords

Aerosol dynamics; general dynamic equation; size distribution function.

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Author Contributions

Author 1: Conceptualization, Methodology, Writing − Original draft preparation. **Author 2**: Writing − Reviewing and Editing. **Author 3**: Writing − Reviewing and Editing.

1. Introduction

A set of approximate equations describing the aerosol dynamics in a uniform control volume (batch reactor) was provided by Friedlander [1] incorporating homogeneous nucleation in terms of the moments of the size distribution of the stable aerosol, $d_p > d_p^*$ and the growth law governed by molecular bombardment (i.e., $d_p < l$). Under such conditions, the calculation of the zeroth, first, and the second moments can be obtained without the determination of the size distribution of the stable particles. In this technical note, the growth law based on diffusion (i.e., $d_p >> l$)[1] is used to derive a similar set of approximate equations. The treatment follows closely to that described in Friedlander [1][2]. The nomenclature also follows that used in Friedlander [1], unless stated otherwise.

[1] Briefly mentioned in Appendix A of the doctoral dissertation of Whitby [2]
[2] On pp. 293-296 in Friedlander [1]

2. Analysis

A situation of homogeneous nucleation and condensation growth in a control volume is examined. Vapor depletion in control volume V (containing air and condensable vapor) is considered. The temperature in the control volume is prescribed as T, a constant.

The general dynamic equation (GDE) for the size distribution function n with ($v >> v_m$) is[3]

$$\frac{\partial n}{\partial t} + \vec{\nabla} \cdot n\vec{V} + \frac{\partial I}{\partial v} = \vec{\nabla} \cdot D\vec{\nabla} n + \frac{1}{2}\int_0^v \beta(\tilde{v}, v - \tilde{v}) n(\tilde{v}) n(v - \tilde{v}) \,\mathrm{d}\tilde{v} - \int_0^v \beta(v, \tilde{v}) n(v) n(\tilde{v}) \,\mathrm{d}\tilde{v} - \vec{\nabla} \cdot \vec{c} n \tag{1}$$

Assuming a spatially uniform $n = n(v,t)$, no coagulation, Brownian diffusion, and sedimentation, Eq. (1) can be simplified to

$$\frac{\partial n}{\partial t} + \frac{\partial I}{\partial v} = 0 \tag{2}$$

Integrating Eq. (2) with respect to v, the dynamic equation for the zeroth moment (total number concentration, N_∞) is obtained. The mathematical manipulation is described in detail in Friedlander [1] [4].

$$\frac{\mathrm{d}N_\infty}{\mathrm{d}t} = I_d \tag{3}$$

The volume fraction ϕ can be obtained by multiplying Eq. (1) by v and integrating with respect to v. The mathematical detail can be found in Friedlander [1][5].

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = \int_{v^*}^{\infty} I dv + \left[Iv \right]_{v^*} \tag{4}$$

To evaluate the first term on the right-hand side of Eq. (4), the following approximation is used[6].

$$I = n\frac{\mathrm{d}v}{\mathrm{d}t} \tag{5}$$

[3] Equation (11.11) on p. 309 in Friedlander [1]
[4] On pp. 310-311 in Friedlander [1]
[5] On pp. 311-312 in Friedlander [1]
[6] On p. 309 in Friedlander [1]

Substituting Eq. (5) into Eq. (4) yields,

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = \int_{v^*}^{\infty} n \frac{\mathrm{d}v}{\mathrm{d}t} dv + \left[Iv \right]_{v^*} \tag{6}$$

The second term on the right-hand side of Eq. (6) can be written as

$$\left[Iv \right]_{v^*} = I_d v^* \tag{7}$$

where v^* is given by

$$v^* = \frac{\pi (d_p^*)^3}{6} \tag{8}$$

with d_p^* given by[7]

$$d_p^* = \frac{4\sigma v_m}{kT \ln S} \tag{9}$$

where $S = p_1 / p_s$.

Assuming diffusion-limited growth, the growth rate $\mathrm{d}v / \mathrm{d}t$ can be obtained from Friedlander [1][8].

$$\frac{\mathrm{d}v}{\mathrm{d}t} = \frac{2\pi D d_p v_m (p_1 - p_d)}{kT} \tag{10}$$

To simplify the analysis further, the Kelvin effect is assumed to be negligible.

$$p_d \approx p_s \tag{11}$$

Substituting Eq. (7), Eq. (8), Eq. (9), Eq. (10), and Eq. (11) into Eq. (6) yields

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = \int_{v^*}^{\infty} n \frac{2\pi D d_p v_m (p_1 - p_s)}{kT} \mathrm{d}v + I_d v^* \tag{12}$$

Knowing $v = \pi (d_p)^3 / 6$, Eq. (12) can be written in terms of the one-third (1/3) moment as

[7] Eq. (9.56) on p. 272 in Friedlander [1]
[8] Eq. (10.25) on p. 285 in Friedlander [1]

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = \frac{2\pi D v_m (p_1 - p_s)}{kT}\left[\frac{6}{\pi}\right]^{1/3} M_{1/3} + I_d v^* \quad (13)$$

where

$$M_{1/3} \equiv \int_{v^*}^{\infty} n v^{1/3} \mathrm{d}v \quad (14)$$

The time rate of change of $M_{1/3}$ is

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t}\left\{\int_{v^*}^{\infty} n v^{1/3} \mathrm{d}v\right\} \quad (15)$$

Using Leibniz's rule and $n \to 0$ as $v \to \infty$, Eq. (15) becomes

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = \int_{v^*}^{\infty} \frac{\partial}{\partial t}\left[n v^{1/3}\right] \mathrm{d}v - \left[n v^{1/3}\right]_{v^*} \frac{\mathrm{d}v^*}{\mathrm{d}t} \quad (16)$$

Assuming that $\mathrm{d}v^* / \mathrm{d}t = 0$, then

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = \int_{v^*}^{\infty} \frac{\partial}{\partial t}\left[n v^{1/3}\right] \mathrm{d}v \quad (17)$$

The term on the right-hand side can be further manipulated using Eq. (2) and Eq. (5).

$$\int_{v^*}^{\infty} \frac{\partial}{\partial t}\left[n v^{1/3}\right] \mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial n}{\partial t} v^{1/3} \mathrm{d}v = -\int_{v^*}^{\infty} \frac{\partial I}{\partial v} v^{1/3} \mathrm{d}v = -\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{1/3} \mathrm{d}v \quad (18)$$

To further evaluate Eq. (18), use is made of the following identity.

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n\, v^{1/3} \frac{\mathrm{d}v}{\mathrm{d}t}\right] \mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{1/3} \mathrm{d}v + \int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[v^{1/3}\right] n \frac{\mathrm{d}v}{\mathrm{d}t} \mathrm{d}v \quad (19)$$

The first term on the right-hand side of Eq. (19) can be expressed as

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n\frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{1/3}\mathrm{d}v = \left[n\,v^{1/3}\,\frac{\mathrm{d}v}{\mathrm{d}t}\right]_{v^*}^{\infty} - \int_{v^*}^{\infty} \frac{1}{3\,v^{2/3}}\,n\frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v \tag{20}$$

Knowing that $n \to 0$ as $v \to \infty$, the first term on the right-hand side of Eq. (20) can be evaluated.

$$\left[n\,v^{1/3}\,\frac{\mathrm{d}v}{\mathrm{d}t}\right]_{v^*}^{\infty} = -\,I_d(v^*)^{1/3} \tag{21}$$

Using Eq. (10), the second term on the right-hand side of Eq. (20) can be written as

$$\int_{v^*}^{\infty} \frac{1}{3\,v^{2/3}}\,n\frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v = \int_{v^*}^{\infty} \frac{1}{3\,v^{2/3}}\,n\frac{2\pi D v_m(p_1 - p_s)}{kT}\left[\frac{6\,v}{\pi}\right]^{1/3}\mathrm{d}v \tag{22}$$

or

$$\int_{v^*}^{\infty} \frac{1}{3\,v^{2/3}}\,n\frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v = \frac{2\pi D v_m(p_1 - p_s)}{3\,kT}\left[\frac{6}{\pi}\right]^{1/3}\int_{v^*}^{\infty} \frac{1}{v^{1/3}}\,n\mathrm{d}v \tag{23}$$

Substituting Eq. (21) and Eq. (23) into Eq. (20) yields

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n\frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{1/3}\mathrm{d}v = -\,I_d(v^*)^{1/3} - \frac{2\pi D v_m(p_1 - p_s)}{3kT}\left[\frac{6}{\pi}\right]^{1/3}\int_{v^*}^{\infty} \frac{1}{v^{1/3}}\,n\,\mathrm{d}v \tag{24}$$

Using Eq. (18) and Eq. (24), Eq. (17) becomes

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = I_d(v^*)^{1/3} + \frac{2\pi D v_m(p_1 - p_s)}{3kT}\left[\frac{6}{\pi}\right]^{1/3}\int_{v^*}^{\infty} \frac{1}{v^{1/3}}\,n\,\mathrm{d}v \tag{25}$$

Define the (–1/3)th moment as

$$M_{-1/3} \equiv \int_{v^*}^{\infty} \frac{1}{v^{1/3}}\,n\,\mathrm{d}v \tag{26}$$

Substituting Eq. (26) into Eq. (25),

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = I_d (v^*)^{1/3} + \frac{2\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} M_{-1/3} \tag{27}$$

To evaluate $M_{-1/3}$ of Eq. (27), Leibniz's rule is used. With $n \to 0$ as $v \to \infty$,

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{v^*}^{\infty} \frac{1}{v^{1/3}} n \mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial}{\partial t} \left[\frac{n}{v^{1/3}}\right] \mathrm{d}v - \left[\frac{n}{v^{1/3}}\right]_{v^*} \frac{\mathrm{d}v^*}{\mathrm{d}t} \tag{28}$$

Again, assuming that $\mathrm{d}v^* / \mathrm{d}t = 0$, Eq. (28) becomes

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{v^*}^{\infty} \frac{1}{v^{1/3}} n \mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial}{\partial t} \left[\frac{n}{v^{1/3}}\right] \mathrm{d}v \tag{29}$$

Using Eq. (2) and Eq. (5), the term on the right-hand side of Eq. (29) can be further simplified as

$$\int_{v^*}^{\infty} \frac{\partial}{\partial t} \left[\frac{n}{v^{1/3}}\right] \mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial n}{\partial t} \left[\frac{1}{v^{1/3}}\right] \mathrm{d}v = -\int_{v^*}^{\infty} \frac{\partial I}{\partial v} \left[\frac{1}{v^{1/3}}\right] \mathrm{d}v = -\int_{v^*}^{\infty} \frac{\partial}{\partial v} \left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] \frac{\mathrm{d}v}{v^{1/3}} \tag{30}$$

To further evaluate Eq. (30), use is made of the following identity.

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v} \left[\frac{n}{v^{1/3}} \frac{\mathrm{d}v}{\mathrm{d}t}\right] \mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial}{\partial v} \left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] \frac{\mathrm{d}v}{v^{1/3}} + \int_{v^*}^{\infty} \frac{\partial}{\partial v} \left[\frac{1}{v^{1/3}}\right] n \frac{\mathrm{d}v}{\mathrm{d}t} \mathrm{d}v \tag{31}$$

The first term on the right-hand side of Eq. (31) can be expressed as

$$\begin{aligned}
\int_{v^*}^{\infty} \frac{\partial}{\partial v} \left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] \frac{\mathrm{d}v}{v^{1/3}} &= \left[\frac{n}{v^{1/3}} \frac{\mathrm{d}v}{\mathrm{d}t}\right]_{v^*}^{\infty} - \int_{v^*}^{\infty} \frac{\partial}{\partial v} \left[\frac{1}{v^{1/3}}\right] n \frac{\mathrm{d}v}{\mathrm{d}t} \mathrm{d}v \\
&= -I_d \left(\frac{1}{v^*}\right)^{1/3} + \frac{1}{3} \int_{v^*}^{\infty} \left[\frac{1}{v^{4/3}}\right] n \frac{\mathrm{d}v}{\mathrm{d}t} \mathrm{d}v \\
&= -I_d \left(\frac{1}{v^*}\right)^{1/3} + \frac{1}{3} \int_{v^*}^{\infty} \left[\frac{1}{v^{4/3}}\right] n \frac{2\pi D v_m (p_1 - p_s)}{kT} \left[\frac{6v}{\pi}\right]^{1/3} \mathrm{d}v \\
&= -I_d \left(\frac{1}{v^*}\right)^{1/3} + \frac{2\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} \int_{v^*}^{\infty} \left[\frac{1}{v}\right] n \mathrm{d}v
\end{aligned} \tag{32}$$

Using Eq. (30) and Eq. (32), Eq. (29) becomes

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_{v^*}^{\infty}\frac{1}{v^{1/3}}n\mathrm{d}v = I_d(\frac{1}{v^*})^{1/3} - \frac{2\pi D v_m(p_1 - p_s)}{3kT}\left[\frac{6}{\pi}\right]^{1/3}\int_{v^*}^{\infty}\left[\frac{1}{v}\right]n\mathrm{d}v \tag{33}$$

Define the (–1)st moment as

$$M_{-1} \equiv \int_{v^*}^{\infty}\frac{1}{v}n\mathrm{d}v \tag{34}$$

Using Eq. (26) and Eq. (34), Eq. (33) becomes

$$\frac{\mathrm{d}M_{-1/3}}{\mathrm{d}t} = I_d(\frac{1}{v^*})^{1/3} - \frac{2\pi D v_m(p_1 - p_s)}{3kT}\left[\frac{6}{\pi}\right]^{1/3} M_{-1} \tag{35}$$

To obtain the governing equation for M_{-1}, the time derivative of Eq. (34) is carried out with Leibniz's rule knowing that $n \to 0$ as $v \to \infty$.

$$\frac{\mathrm{d}M_{-1}}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t}\int_{v^*}^{\infty}\frac{1}{v}n\mathrm{d}v = \int_{v^*}^{\infty}\frac{\partial}{\partial t}\left[\frac{n}{v}\right]\mathrm{d}v - \left[\frac{n}{v}\right]_{v^*}\frac{\mathrm{d}v^*}{\mathrm{d}t} \tag{36}$$

Again, assuming $\mathrm{d}v^* / \mathrm{d}t = 0$ and using Eq. (2) and Eq. (5), Eq. (36) becomes

$$\begin{aligned}\frac{\mathrm{d}M_{-1}}{\mathrm{d}t} &= \frac{\mathrm{d}}{\mathrm{d}t}\int_{v^*}^{\infty}\frac{1}{v}n\mathrm{d}v = \int_{v^*}^{\infty}\frac{\partial}{\partial t}\left[\frac{n}{v}\right]\mathrm{d}v = \int_{v^*}^{\infty}\frac{\partial n}{\partial t}\frac{1}{v}\mathrm{d}v \\ &= -\int_{v^*}^{\infty}\frac{\partial I}{\partial v}\frac{1}{v}\mathrm{d}v = -\int_{v^*}^{\infty}\frac{\partial}{\partial v}\left[n\frac{\mathrm{d}v}{\mathrm{d}t}\right]\frac{1}{v}\mathrm{d}v\end{aligned} \tag{37}$$

To further evaluate Eq. (37), use is made of the following identity.

$$\int_{v^*}^{\infty}\frac{\partial}{\partial v}\left[\frac{n}{v}\frac{\mathrm{d}v}{\mathrm{d}t}\right]\mathrm{d}v = \int_{v^*}^{\infty}\frac{\partial}{\partial v}\left[n\frac{\mathrm{d}v}{\mathrm{d}t}\right]\frac{\mathrm{d}v}{v} + \int_{v^*}^{\infty}\frac{\partial}{\partial v}\left[\frac{1}{v}\right]n\frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v \tag{38}$$

From Eq. (38) and using Eq. (10),

$$\begin{aligned}\int_{v^*}^{\infty}\frac{\partial}{\partial v}\left[n\frac{\mathrm{d}v}{\mathrm{d}t}\right]\frac{\mathrm{d}v}{v} &= \left[\frac{n}{v}\frac{\mathrm{d}v}{\mathrm{d}t}\right]_{v*}^{\infty} - \int_{v^*}^{\infty}\frac{\partial}{\partial v}\left[\frac{1}{v}\right]n\frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v \\ &= -\frac{I_d}{v^*} + \int_{v^*}^{\infty}\left[\frac{1}{v^2}\right]n\frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v \\ &= -\frac{I_d}{v^*} + \int_{v^*}^{\infty}\left[\frac{1}{v^2}\right]n\frac{2\pi D v_m(p_1 - p_s)}{kT}\left[\frac{6v}{\pi}\right]^{1/3}\mathrm{d}v \\ &= -\frac{I_d}{v^*} + \frac{2\pi D v_m(p_1 - p_s)}{kT}\left[\frac{6}{\pi}\right]^{1/3}\int_{v^*}^{\infty}\left[\frac{n}{v^{5/3}}\right]\mathrm{d}v\end{aligned} \tag{39}$$

Define the (–5/3)rd moment as

$$M_{-5/3} \equiv \int_{v^*}^{\infty}\frac{1}{v^{5/3}}n\mathrm{d}v \tag{40}$$

Using Eq. (39) and Eq. (40), Eq. (37) becomes

$$\frac{\mathrm{d}M_{-1}}{\mathrm{d}t} = \frac{I_d}{v^*} - \frac{2\pi D v_m(p_1 - p_s)}{kT}\left[\frac{6}{\pi}\right]^{1/3} M_{-5/3} \tag{41}$$

Similarly, one can derive the recursion formula for the ($-j\,/\,3$)rd moment with $j = 1, 3, 5, \cdots$.

$$\frac{\mathrm{d}M_{-j/3}}{\mathrm{d}t} = I_d\left[\frac{1}{v^*}\right]^{j/3} - \frac{j2\pi D v_m(p_1 - p_s)}{3kT}\left[\frac{6}{\pi}\right]^{1/3} M_{-(j+2/3)} \tag{42}$$

In general, we can define

$$\forall k \in \mathbb{R} \qquad M_{-k} \equiv \int_{v^*}^{\infty}\frac{1}{v^k}n\mathrm{d}v \tag{43}$$

From Eq. (13), the total aerosol mass, M_p, in the control volume V at any time t is

$$\frac{\mathrm{d}M_p}{\mathrm{d}t} = \rho_p V\frac{\mathrm{d}\phi}{\mathrm{d}t} = \rho_p V\left\{\frac{2\pi D v_m(p_1 - p_s)}{kT}\left[\frac{6}{\pi}\right]^{1/3} M_{1/3} + I_d\, v^*\right\} \tag{44}$$

Equation (44) can be written in terms of C.

$$\frac{\mathrm{d}C}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{M_p}{V}\right] = \rho_p \left\{\frac{2\pi D v_m (p_1 - p_s)}{kT}\left[\frac{6}{\pi}\right]^{1/3} M_{1/3} + I_d v^*\right\} \tag{45}$$

Vapor mass (M_v) depletion can be calculated by

$$-\frac{\mathrm{d}M_v}{\mathrm{d}t} = \frac{\mathrm{d}M_p}{\mathrm{d}t} \tag{46}$$

or

$$\frac{\mathrm{d}Y_v}{\mathrm{d}t} = \frac{RT}{P_{total}VM}\frac{\mathrm{d}M_v}{\mathrm{d}t} = -\frac{RT}{P_{total}M}\frac{\mathrm{d}C}{\mathrm{d}t} \tag{47}$$

Equation (47) can be written in terms of p_1 as follow.

$$\frac{\mathrm{d}p_1}{\mathrm{d}t} = \frac{RT}{VM}\frac{\mathrm{d}M_v}{\mathrm{d}t} = -\frac{RT}{M}\frac{\mathrm{d}C}{\mathrm{d}t} \tag{48}$$

or

$$\frac{\mathrm{d}S}{\mathrm{d}t} \equiv \frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{p_1}{p_s}\right] = \frac{RT}{p_s VM}\frac{\mathrm{d}M_v}{\mathrm{d}t} = -\frac{RT}{p_s M}\frac{\mathrm{d}C}{\mathrm{d}t} \tag{49}$$

Substituting Eq. (45) into Eq. (49),

$$\frac{\mathrm{d}S}{\mathrm{d}t} = -\frac{RT}{M}\rho_p \left\{\frac{2\pi D v_m (S-1)}{kT}\left[\frac{6}{\pi}\right]^{1/3} M_{1/3} + \frac{I_d v^*}{p_s}\right\} \tag{50}$$

The (2/3)rd moment is defined as

$$M_{2/3} = \int_{v^*}^{\infty} n v^{2/3} \mathrm{d}v \tag{51}$$

The time rate of change of $M_{2/3}$ is

$$\frac{\mathrm{d}M_{2/3}}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t}\left\{\int_{v^*}^{\infty} n\, v^{2/3} \mathrm{d}v\right\} \tag{52}$$

Using Leibniz's rule with $n \ \to 0$ as $v \ \infty \to$, Eq. (52) becomes

$$\frac{\mathrm{d}M_{2/3}}{\mathrm{d}t} = \int_{v^*}^{\infty} \frac{\partial}{\partial t}\left[n\, v^{2/3}\right]\mathrm{d}v - \left[n\, v^{2/3}\right]_{v^*} \frac{\mathrm{d}v^*}{\mathrm{d}t} \tag{53}$$

Assuming that $\mathrm{d}v^* \,/\, \mathrm{d}t = 0$, then

$$\frac{\mathrm{d}M_{2/3}}{\mathrm{d}t} = \int_{v^*}^{\infty} \frac{\partial}{\partial t}\left[n v^{2/3}\right]\mathrm{d}v \tag{54}$$

The term on the right-hand side can be further manipulated using Eq. (2) and Eq. (5).

$$\int_{v^*}^{\infty} \frac{\partial}{\partial t}\left[n v^{2/3}\right]\mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial n}{\partial t} v^{2/3}\mathrm{d}v = -\int_{v^*}^{\infty} \frac{\partial I}{\partial v} v^{2/3}\mathrm{d}v = -\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{2/3}\mathrm{d}v \tag{55}$$

To further evaluate Eq. (55), use is made of the following identity.

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n v^{2/3} \frac{\mathrm{d}v}{\mathrm{d}t}\right]\mathrm{d}v = \int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{2/3}\mathrm{d}v + \int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[v^{2/3}\right] n \frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v \tag{56}$$

From Eq. (56),

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{2/3}\mathrm{d}v = \left[n\, v^{2/3} \frac{\mathrm{d}v}{\mathrm{d}t}\right]_{v^*}^{\infty} - \int_{v^*}^{\infty} \frac{2}{3\, v^{1/3}}\, n \frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v \tag{57}$$

Knowing that $n \to 0$ as $v \to \infty$, the first term on the right-hand side of Eq. (57) can be evaluated.

$$\left[n v^{2/3} \frac{\mathrm{d}v}{\mathrm{d}t}\right]_{v^*}^{\infty} = -\, I_d (v^*)^{2/3} \tag{58}$$

Using Eq. (10), the second term on the right-hand side of Eq. (57) can be written as

$$\int_{v^*}^{\infty} \frac{2}{3v^{1/3}}\, n \frac{\mathrm{d}v}{\mathrm{d}t}\mathrm{d}v = \int_{v^*}^{\infty} \frac{2}{3v^{1/3}}\, n \frac{2\pi D v_m (p_1 - p_s)}{kT} \left[\frac{6\, v}{\pi}\right]^{1/3} \mathrm{d}v \tag{59}$$

or

$$\int_{v^*}^{\infty} \frac{2}{3v^{1/3}} n \frac{\mathrm{d}v}{\mathrm{d}t} \mathrm{d}v = \frac{4\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} \int_{v^*}^{\infty} n \mathrm{d}v \tag{60}$$

Substituting Eq. (58) and Eq. (60) into Eq. (57) yields

$$\int_{v^*}^{\infty} \frac{\partial}{\partial v}\left[n \frac{\mathrm{d}v}{\mathrm{d}t}\right] v^{2/3} \mathrm{d}v = -I_d (v^*)^{2/3} - \frac{4\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} \int_{v^*}^{\infty} n \mathrm{d}v \tag{61}$$

Using Eq. (55) and Eq. (61), Eq. (54) becomes

$$\frac{\mathrm{d}M_{2/3}}{\mathrm{d}t} = I_d (v^*)^{2/3} + \frac{4\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} N_\infty \tag{62}$$

Based on the above analysis, the following set of differential equations describes the homogeneous nucleation and diffusional growth processes in a uniform control volume.

$$\frac{\mathrm{d}N_\infty}{\mathrm{d}t} = I_d \tag{3}$$

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = I_d (v^*)^{1/3} + \frac{2\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} M_{-1/3} \tag{27}$$

$$\frac{\mathrm{d}M_{-j/3}}{\mathrm{d}t} = I_d \left[\frac{1}{v^*}\right]^{j/3} - \frac{j 2\pi D v_m (p_1 - p_s)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} M_{-\left(j+2/3\right)} \quad \text{for } j = 1, 3, 5, \cdots \tag{42}$$

$$\frac{\mathrm{d}M_{2/3}}{\mathrm{d}t} = I_d (v^*)^{2/3} + \frac{4\pi D v_m p_s (S-1)}{3kT} \left[\frac{6}{\pi}\right]^{1/3} N_\infty \tag{62}$$

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = I_d v^* + \frac{2\pi D v_m p_s (S-1)}{kT} \left[\frac{6}{\pi}\right]^{1/3} M_{1/3} \tag{13}$$

$$\frac{\mathrm{d}S}{\mathrm{d}t} = -\frac{RT}{M} \rho_p \left\{ \frac{2\pi D v_m (S-1)}{kT} \left[\frac{6}{\pi}\right]^{1/3} M_{1/3} + \frac{I_d v^*}{p_s} \right\} \tag{50}$$

with $I_d = I_d(S)$ given in Friedlander [1][9] and v^* given by Eq. (8) above. However, due to the

recursive nature of Eq. (42), it is impossible to obtain closure for the moment equations.

One way to address the closure problem is to specify a size distribution function. As an example, a lognormal distribution is assumed and given by

$$n(v,t) = \frac{N_\infty(t)}{\sqrt{2\pi}\, v\, \sigma_n(t)} \exp\left\{-\frac{1}{2}\left[\frac{\left(\ln v - \ln v_n(t)\right)}{\sigma_n(t)}\right]^2\right\} \tag{63}$$

The j^{th} moment of a lognormal distribution function can be obtained from Crow and Shimizu [3] and is given below.

$$M_j = N_\infty \exp\left[j \ln v_n + \frac{1}{2}(j\sigma_n)^2\right] \tag{64}$$

From Eq. (64), the first moment $M_1 = \phi$ and the (1/3)rd moment $M_{1/3}$ are given by

$$M_1 = N_\infty \exp\left[\ln v_n + \frac{1}{2}(\sigma_n)^2\right] \tag{65}$$

$$M_{1/3} = N_\infty \exp\left[\frac{1}{3}\ln v_n + \frac{1}{2}(\frac{1}{3}\sigma_n)^2\right] \tag{66}$$

From Eq. (65) and Eq. (66), the following relationships are obtained.

$$v_n = \frac{(M_{1/3})^{9/2}}{\sqrt{M_1 N_\infty^4}} = \frac{(M_{1/3})^{9/2}}{\sqrt{\phi\, N_\infty^4}} \tag{67}$$

$$\sigma_n{}^2 = \ln\left[\frac{M_1^3 N_\infty^6}{(M_{1/3})^9}\right] = \ln\left[\frac{\phi^3 N_\infty^6}{(M_{1/3})^9}\right] \tag{68}$$

The governing equations for the problem become

$$\frac{\mathrm{d}N_\infty}{\mathrm{d}t} = I_d \tag{3}$$

[9] Eq. (10.16) on p. 279 in Friedlander [1]

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = I_d v^* + \frac{2\pi D v_m p_s (S-1)}{kT} \left[\frac{6}{\pi}\right]^{1/3} M_{1/3} \tag{13}$$

$$\frac{\mathrm{d}S}{\mathrm{d}t} = -\frac{RT}{M}\rho_p \left\{\frac{2\pi D v_m (S-1)}{kT}\left[\frac{6}{\pi}\right]^{1/3} M_{1/3} + \frac{I_d v^*}{p_s}\right\} \tag{50}$$

$$\frac{\mathrm{d}M_{1/3}}{\mathrm{d}t} = I_d (v^*)^{1/3} + \frac{2\pi D v_m (p_1 - p_s)}{3kT}\left[\frac{6}{\pi}\right]^{1/3} M_{-1/3} \tag{27}$$

$$M_{-1/3} = N_\infty \exp\left[-\frac{1}{3}\ln v_n + \frac{1}{2}(-\frac{1}{3}\sigma_n)^2\right] \tag{69}$$

$$v_n = \frac{(M_{1/3})^{9/2}}{\sqrt{M_1} N_\infty^4} = \frac{(M_{1/3})^{9/2}}{\sqrt{\phi}\, N_\infty^4} \tag{67}$$

$$\sigma_n{}^2 = \ln\left[\frac{M_1^3 N_\infty^6}{(M_{1/3})^9}\right] = \ln\left[\frac{\phi^3 N_\infty^6}{(M_{1/3})^9}\right] \tag{68}$$

3. Concluding Remarks

In this technical note, a set of approximate first-order ordinary differential equations was derived to study the aerosol dynamics with homogeneous nucleation and condensational diffusion-limited growth in an isothermal batch reactor. A lognormal size distribution was used to demonstrate how to address the closure problem associated with the recursive moment equations in the formulation. The resulting set of approximate equations can be solved numerically to obtain the temporal variations of number concentration, volume fraction, vapor depletion, and other moments.

References

[1] Friedlander SK (2000) *Smoke, Dust, and Haze. Fundamentals of Aerosol Dynamics* (Oxford University Press, New York), 2nd Ed.

[2] Whitby ER (1990) Modal Aerosol Dynamics Modeling. Ph.D. Thesis. (University of Minnesota).

[3] Crow EL and Shimizu K (eds.) (1988) *Lognormal Distributions, Theory and Applications* (Marcel Dekker, Inc. New York).

Appendix A. List of Symbols

$\vec{c}$
Migration velocity in the presence of an external force field

C
Particle mass concentration

d_p
Particle diameter

d_p^*
Critical nucleus size

D
Diffusion coefficient of condensing species

I
Particle current

I_d
Homogeneous nucleation rate

k
Boltzmann's constant

l
Mean free path of condensing species

m
Molecular mass

M
Molecular weight of condensing vapor

M_j
Moments of particle size distribution function

M_p
Total aerosol mass

M_v
Condensable vapor mass

$n(\cdots)$
Particle volume distribution function

N_∞
Total particle number concentration

p_1
Monomer partial pressure

p_d

Equilibrium vapor pressure above a drop of diameter d_p

p_s

Equilibrium vapor pressure above a planar surface

R

Universal gas constant

S

Saturation ratio

t

Time

T

Absolute temperature

$v, \tilde{v}$

Particle volume

v_m

Molecular volume of condensing species

v_n

Lognormal distribution parameter

v^*

Critical particle volume

V

Volume of batch reactor

$\vec{V}$

Particle velocity

Y_v

Amount of condensable vapor fraction

β

Collision frequency function

ϕ

Aerosol volume fraction

σ

Surface tension

σ_n

Lognormal distribution parameter